

**REMARKS**

Claims 1 to 35 are all the claims pending in the application, prior to the present amendment.

Claims 4-6, 9-22, 24, 28 32 and 33 have been objected to as being improper multiple dependent claims because a multiple dependent claim may not depend from another multiple dependent claim. The Examiner states that the claims have been treated on the merits, but the failure to correct this deficiency will result in the claims being removed from consideration.

In response, applicants have amended the claims as set forth above so that the claims no longer contain improper multiple dependencies.

Accordingly, applicants request withdrawal of this objection.

Claims 22, 23, 25-33 and 35 have been rejected under 35 U.S.C. § 102(b) as anticipated by the Wu et al article in *Chemistry Letters* 32:774-775 (2000).

Applicants have canceled claims 22, 23 and 25 to 32, thus leaving only claims 33 and 35 as being subject to this rejection.

Applicants submit that the products of claims 33 and 35 are patentable over Wu et al, since the processes of claims 1 and 34, respectively, from which these claims depend are novel and inventive and, thus, the products of these processes are novel and inventive.

For a better understanding, applicants set forth below a detailed explanation regarding the differences in process and product between the present invention and Wu et al.

**(1) Characteristic feature of MWW-type zeolite**

In the case of a substance generally called a zeolite, one can obtain a product with a zeolite structure having a three-dimensional network directly by hydrothermal synthesis, while

in the case of a zeolite having an MWW structure, the product obtained by hydrothermal synthesis is a layered precursor having a two-dimensional layered structure and one has to calcine the precursor to convert it into a zeolite having a three-dimensional network. The present invention has been completed advantageously utilizing a novel synthesis procedure of an MWW-type zeolite.

(2) Conventional process of Wu et al

The conventional process disclosed in the cited Wu et al reference comprises the following three steps.

First step: preparation of precursor

The starting materials consisting of a B source, a Ti source, an Si source, water and a template compound are reacted under hydrothermal condition to obtain a layered precursor in which B and Ti are incorporated into the framework.

Second step: removal of boron by acid treatment

The precursor obtained in the first step (or the precursor additionally calcined) is acid treated to remove boron. At this time, it is necessary to select appropriate conditions for obtaining a product with enough Ti content and less B content, since boron cannot be substantially completely removed unless severe conditions are employed, but Ti is also removed under such severe conditions.

Third step: conversion into zeolite by calcining

The substance obtained in the second step is calcined to completely burn and remove the template compound and form a three-dimensional network, thereby obtaining an MWW-type zeolite.

(3) Process of the invention

The process of the present invention for producing a zeolite substance as set forth in claim 1 comprises at least the following four steps.

First step: preparation of precursor

The starting materials consisting of a B source, an Si source, water and a template compound are reacted under hydrothermal condition to obtain a layered precursor in which B is incorporated into the framework. Elements other than B, such as Ti, are not incorporated at this time.

Second step: removal of boron by acid treatment

The precursor obtained in the first step (or the precursor additionally calcined) is acid treated to remove boron. As a result, a layered precursor is formed in which the sites at which boron existed have become defective.

Boron can be substantially completely removed, since severe conditions can be employed.

Third step: second hydrothermal synthesis in presence of heteroatoms such as Ti

The layered precursor obtained in the second step is mixed with a compound containing a metal element such as Ti, water and a template compound and again reacted under hydrothermal condition to obtain a layered precursor in which the template compound exists in the inter layer spaces. At this time, the metal element is incorporated into the defect sites at which B existed so that the precursor is converted into a metallosilicate layered precursor.

Fourth step: conversion into zeolite by calcining

The substance obtained in the third step is calcined to remove the template compound and form a three-dimensional network, thereby obtaining an MWW-type zeolite.

The process of the present invention for producing a layered precursor for a zeolite substance comprises the above first to third steps.

**(4) Novelty of the invention**

The inventors of the present invention believe that there has not hitherto been known a process in which a layered precursor of MWW-type zeolite is once synthesized, then defect sites are formed in the precursor, and thereafter hydrothermal synthesis is again performed in the presence of a metal compound to obtain a layered precursor in which a metal element is incorporated, as in the process according to the present invention.

In the process of the present invention, a layered precursor of MWW-type zeolite having defect sites is synthesized, and then the inter layer spaces of the precursor are enlarged by the template compound and contacted with the metal compound so that the precursor can be converted into a metallosilicate having a layered structure in which a metal element is effectively incorporated into the defect sites.

**(5) Comparison of two processes**

For further understanding, applicants enclose herewith schematic drawings illustrating the production process for an MWW-type metallosilicate by the conventional hydrothermal synthesis method described in Wu et al and the production process for an MWW-type metallosilicate by the new hydrothermal synthesis method according to the present invention.

Further, a comparison in effect between the process of the present invention and the conventional process of Wu et al is given below,

a. **Metal species which can be introduced into the framework**

In the process of the present invention, metal elements having a large ionic radius which cannot be introduced into the framework by the conventional hydrothermal synthesis method, even metal elements in Period 4 or more of the periodic table, can effectively be introduced, since the metal element is introduced after the formation of the defect sites.

On the contrary, in the process of Wu et al, a metal element and B should both exist for obtaining the precursor of MWW-type zeolite, and therefore B having a small ionic radius is easily selectively introduced into the framework. It is difficult to introduce large ionic radius metal elements in the Period 4 or more of the periodic table into the framework. Although metal elements having an ionic radius analogous to that of Ti can be introduced, it is necessary to feed a large excessive amount of the metal compound.

b. **Removal of B**

In the process of the present invention, B can be substantially completely removed, since the acid treatment is performed in the state that the metal element such as Ti has not yet been introduced.

Contrary to this, in the process of Wu et al, B must be caused to be retained in order that Ti is retained to some extent, since Ti is also removed if the removal of B is enhanced.

c. **Crystallographic site into which metal element is introduced**

In the process of the present invention, the metal element is to exist at a position at which B is easily introduced at the time of synthesis of the layered precursor, since the metal

element is introduced into the defect sites formed by the removal of B. At present, it is supposed that the metal element is selectively introduced into a position that faces the outer surface of the layered compound. Therefore, it is considered that high catalytic activity is attained, due to the fact that the metal element is easily contacted with the reactants in the catalytic reaction or the like.

On the contrary, in the process of Wu et al, Ti is to be introduced into a position at which B is difficult to be introduced, since the hydrothermal synthesis is performed in the state that both B and Ti exist. At present, it is supposed that Ti is mainly introduced into the inside of the layered compound.

As can be seen from the above discussion, the zeolite substance of claim 33 produced by the process of claim 1 and the layered precursor of claim 35 produced by the process of claim 34 are different from the products obtained by the Wu et al process.

In view of the above, applicants submit that Wu et al do not disclose or render obvious the subject matter of claims 33 and 35 and, accordingly, request withdrawal of this rejection.

Claims 1-35 have been provisionally rejected on the ground of nonstatutory obviousness-type double patenting as being unpatentable over claims 1-27 of copending application no. 10/506,366.

In response, applicants enclose herewith a Submission of Terminal Disclaimer, an executed Terminal Disclaimer and the appropriate fee.

Applicants request withdrawal of this rejection in view of the executed Terminal Disclaimer.

In view of the above, reconsideration and allowance of this application are now believed to be in order, and such actions are hereby solicited. If any points remain in issue which the Examiner feels may be best resolved through a personal or telephone interview, the Examiner is kindly requested to contact the undersigned at the telephone number listed below.

The USPTO is directed and authorized to charge all required fees, except for the Issue Fee and the Publication Fee, to Deposit Account No. 19-4880. Please also credit any overpayments to said Deposit Account.

Respectfully submitted,

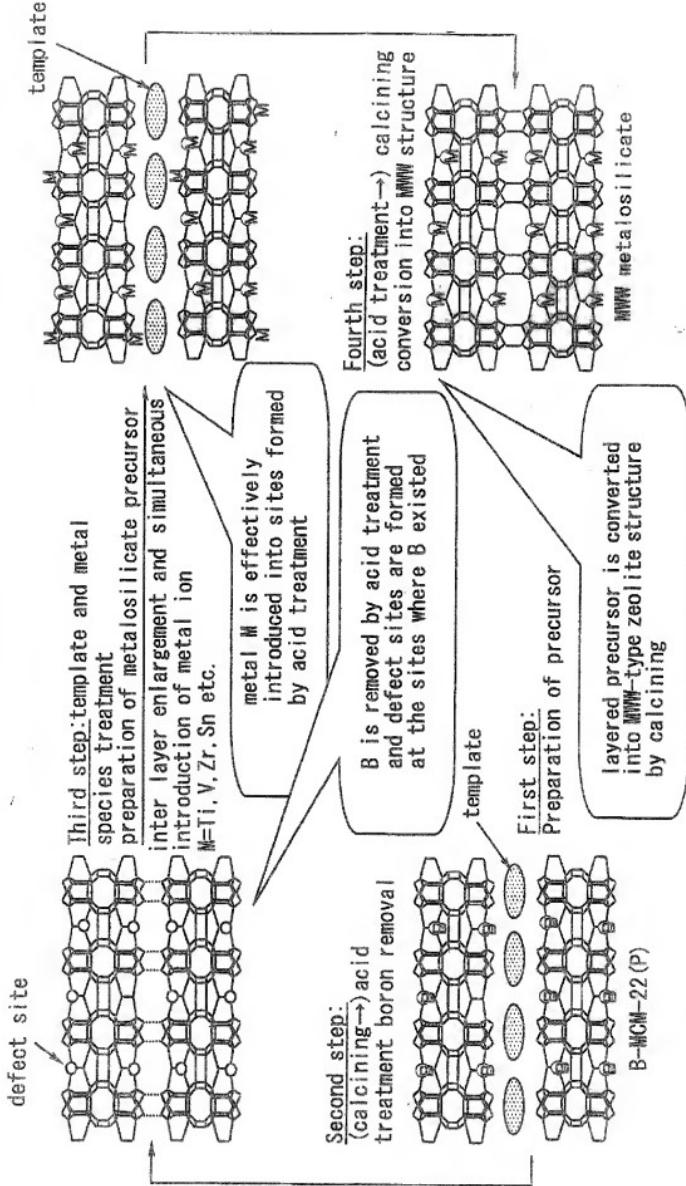
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Date: June 27, 2007

## Production Process for **WWW**-type Metalosilicate According to Present Invention



## Production Process for MWW-type Metasilicate According to Wu et al

